#### [JP-A-08-283708]

# ORGANIC ELECTROLUMINESCENT ELEMENT AND ELECTROPHOTOGRAPHIC PHOTORECEPTOR

#### \* NOTICES \*

JPO and INPIT are not responsible for any damages caused by the use of this translation.

- 1. This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.\*\*\* shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

#### [Claims]

[Claim 1] This order comes to laminate a cathode, a luminous layer, a glue line, and an anode at least on a base substance, and said luminous layer is formula (I).

#### [Chemical formula 1]

$$R^{2}$$
  $N-Ar^{1}-CH=CH-Ar-CH=CH-Ar^{1}-N$   $R^{1}$   $R^{2}$   $R^{3}$   $C=CH$   $R^{3}$   $R^{2}$   $R^{3}$ 

the inside of a formula, and Ar -- a methylene group or an allylene group of the carbon numbers 6-20 which may be replaced. An allylene group of the carbon numbers 6-20 by which Ar<sup>1</sup> may be replaced, A heterocycle group which forms a low-grade alkyl group, a five-membered ring, or

six membered-rings by which R<sup>1</sup> may be replaced, An aralkyl group which may be replaced, or an aryl group of the carbon numbers 6-14 which may be replaced, R<sup>2</sup> and R<sup>3</sup> express a hydrogen atom, a low-grade alkyl group, or an aryl group of the carbon numbers 6-14 that may be replaced. An organic electroluminescence element containing a distilbene compound shown.

[Claim 2] A distilbene compound of formula (I) is formula (II).

#### [Chemical formula 2]

(a hydrogen atom, a halogen atom, a low-grade alkyl group or a lower alkoxy group, and n of R<sup>4</sup> are the integers of 1 or 2 among a formula.) -- the organic electroluminescence element according to claim 1 which is a compound shown.

[Claim 3] The organic electroluminescence element according to claim 1 by which a hole injection layer is further formed between a cathode and a luminous layer.

[Claim 4] It has a photosensitive layer which laminates a charge generating layer and a charge transport layer in this order on a conductive substrate, and said charge transport layer is formula (I).

#### [Chemical formula 3]

the inside of a formula, and Ar -- a methylene group or an allylene group of the carbon numbers 6-20 which may be replaced. An allylene group of the carbon numbers 6-20 by which Ar<sup>1</sup> may be replaced, A heterocycle group which forms a low-grade alkyl group, a five-membered ring, or six membered-rings by which R<sup>1</sup> may be replaced, An aralkyl group which may be replaced, or an aryl group of the carbon numbers 6-14 which may be replaced, R<sup>2</sup> and R<sup>3</sup> express a hydrogen atom, a low-grade alkyl group, or an aryl group of the carbon numbers 6-14 that may be replaced. An electrophotographic photoreceptor element containing a distilbene compound shown.

[Claim 5] A distilbene compound of formula (I) is formula (III).

[Chemical formula 4]

(Ar<sup>2</sup> expresses among a formula a methylene group or a phenylene group, a low-grade alkyl group by which R<sup>5</sup> may be replaced, an aralkyl group which may be replaced, or an aryl group of the carbon numbers 6-14 which may be replaced.) -- the electrophotographic photoreceptor element according to claim 4 which is a compound shown.

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the new distilbene compound which can be adapted for an organic electroluminescence element and an electrophotographic photoreceptor element, and its manufacturing method.

[0002]

[Description of the Prior Art] Visibility of an electroluminescent element (EL element) is high because of self-luminescence, and it is a perfect solid state component.

Since it has the feature of excelling in shock resistance, various elements using inorganic matter and an organic compound are proposed, and utilization is tried now.

Among these, since especially the organic EL device can reduce impressed electromotive force substantially, development of various materials and an element is furthered.

[0003] In order that the composition of an organic EL device may raise luminescent ability by making a cathode / luminous layer / anode into basic constitution, providing a hole injection layer, an electronic injection layer, or a hole barrier layer if needed is known. Among these, when forming a hole barrier layer between a luminous layer and an anode, the energy level of a hole barrier layer must use material which differs from the energy level of a luminous layer. For example, to JP,H2-195683,A, the technology of providing the hole blocking layer (hole barrier layer) as for which more than 0.1V has the first big oxidation potential from the first oxidation potential of a luminous layer between a luminous layer and an anode is indicated. To JP,H2-195683,A and JP,H2-255788,A, using a 8-hydroxyquinoline derivative as a hole barrier layer is

indicated.

[0004] However, even when a 8-hydroxyquinoline derivative is used as a hole barrier layer, about luminescence of a blue system, luminous efficiency is as low as 0.3 (1 m-W<sup>-1</sup>). On the other hand, when the material indicated to Tokuganhei2-242669 and Tokuganhei2-279304 is used for a luminous layer, Since high-intensity luminescence in a blue system can be obtained, using effectively towards full-color-izing of a flat-panel display etc. from now on is expected. However, when the element which has composition, such as cathode / luminous layer / anode/, or a cathode / hole injection layer / luminous layer / anode, using such materials is formed, while producing luminescence unevenness and a non-luminous region, the problem to utilization of the life of an element, micro processing, etc. is not yet solved. [0005] about an electrophotographic photoreceptor element, the material which forms a photo conductor is already changing from inorganic system materials, such as Se, ZnO, and CdS, for organic system materials, such as various kinds of organic system paints and a color, -- the -- the following Reasons can be considered if a factor is carried out. That is, organic system material tends to obtain the material according to various uses than the diversity of (1) compound. (2) Even if it does not vapor-deposit, a photo conductor is easily producible with a coating method. (3) There are few problems of safe. (4) Environmental \*\*\*\* is good (there is little especially degradation in the time of high-humidity/temperature). (5) Also in durable \*\*\*\*, it is satisfactory.

(6) High print durability is obtained.

[0006] It is the early stage of having come to use organic system material, and, now, the problem of the sensitivity made especially into the problem is thoroughly solved by using the photo conductor constituted on a conductive substrate by laminating a charge generating layer and a charge transport layer one by one.

The photo conductor with a big number has been called for the whole copy-proof with the photo conductor with a quick copy speed, and the high copy speed by byway-ization especially in recent years.

[0007] The electrophotographic photoreceptor of a functional discrete type not only can choose a desirable material from the wide range on manufacture, but has the advantage that the width of selection of the material which can raise photosensitivity and endurance is wide. for example, -- as a concrete organic system material -- an azo pigment, BERIREN paints, polycyclic quinone paints, and indigoid paints (JP,54-139540,A.) 55-88064, 56-4148, 56-14240, A pyrrolo pyrrole compound (USP No. 4632893) is mentioned further 56-119131, 63-935455, 63-63046,

Tokuganhei1-109352, USP No. 4952472, DE-2237680, the 2734288 No., etc.

[0008] An oxazol compound meltable to toluene, dichloroethane, or a methylene chloride as a charge transport nature substance which constitutes a photo conductor, An oxadiazole compound, a triphenylamine compound, a styryl compound, a hydrazone compound and enamine compounds (JP,H1-236280,A.) JP,H1-226859,A, JP,H3-30133,B, JP,S60-34099,B, Although

JP,H1-261648,A, JP,H2-272571,A, JP,S58-131954,A, JP,H4-101153,A, and JP,H4-149446,A are mentioned, Since it is [ of / most / these ] deficient in film forming ability, binder resin is usually added. As these resin, polycarbonate resin, polyarylate resin, Polyester resin and polysulphone resin are mentioned and it is used a charge transport nature substance, almost tales doses, or mostly about 10%, Binder resin with high (even if thickness thickens, a remarkable sensitivity lowering phenomenon does not happen easily.) transport efficiency, and a charge transport nature substance have been called for strongly that there is little mechanical deterioration.

[0009]

[Means for solving problem] This invention persons found out that the distilbene compound which has a new structure was effective, as a result of compounding and evaluating the organic material corresponding to each SUBJECT of an organic EL device and an electrophotographic photoreceptor element. That is, according to this invention, this order comes to laminate a cathode, a luminous layer, a glue line, and an anode at least on a base substance, and said luminous layer is formula (I).

[0010]

[Chemical formula 5]

$$\begin{array}{c}
R^{1} \\
R^{2} \\
C = CH
\end{array}$$
N-Ar<sup>1</sup>-CH=CH-Ar-CH=CH-Ar<sup>1</sup>-N
CH=C
R<sup>3</sup>
(1)

[0011] the inside of a formula, and Ar -- a methylene group or the allylene group of the carbon numbers 6-20 which may be replaced. The allylene group of the carbon numbers 6-20 by which Ar<sup>1</sup> may be replaced, The heterocycle group which forms the low-grade alkyl group, the five-membered ring, or six membered-rings by which R<sup>1</sup> may be replaced, The aralkyl group which may be replaced, or the aryl group of the carbon numbers 6-14 which may be replaced, R<sup>2</sup> and R<sup>3</sup> express a hydrogen atom, a low-grade alkyl group, or the aryl group of the carbon numbers 6-14 that may be replaced. The organic electroluminescence element containing the distilbene compound shown is provided.

[0012] According to this invention from another viewpoint, it has a photosensitive layer which laminates a charge generating layer and a charge transport layer in this order on a conductive substrate, and the electrophotographic photoreceptor containing the distilbene compound in which said charge transport layer is shown by the above-mentioned formula (I) is provided. Especially as a base substance in the organic EL device of this invention, it is not limited, the thing of various construction material, such as glass, Ceramics Sub-Division, and hard plastics, can be used, for example, and a desirable transparent substrate is mentioned.

[0013] It is preferred to use the large (not less than 4 eV) metal, the alloy, the electrical conductivity compounds, and these mixtures of a work function as an electrode material which forms a cathode. As an example of such an electrode material, dielectric transparent materials, such as metal, such as Au, CuI, ITO, SnO<sub>2</sub>, or ZnO, are mentioned. Although the thickness in

particular of these electrode materials is not limited, it is preferred to choose suitably in 10-200 nm preferably 10 nm - 1 micrometer. When forming a cathode, the above-mentioned electrode material can be produced by publicly known methods, such as vacuum evaporation and sputtering. As for the sheet resistance as a cathode, it is preferred to set up below hundreds of ohms / \*\* become. When taking out luminescence from this electrode, it is preferred to produce using material in which transmissivity becomes larger than 10%.

[0014] As a luminous layer in the organic EL device of this invention, The compound of the above-mentioned formula (I) is contained, and it is formed, and is \*\* pouring function (at the time of voltage impressing.) like the usual luminous layer. an electron hole can be poured in from a cathode or a hole injection layer, and an electron can be poured in from an anode or an electronic injection layer. \*\* While having a transportation function (it is possible to move an electron hole and an electron with the power of electric field.), and \*\* luminescence function (it is possible to provide an electron hole and the place of electronic recombination and to make them emit light.), it has a luminescence function etc. which tie the above-mentioned function to luminescence. An electron hole is poured in, with easy, an electron is poured in and there may be a difference in easy. It is preferred to move either, although the transportation function in which it is expressed with an electron hole and the degree of electron transfer may vary in size.

Although the thickness in particular of a luminous layer is not limited, it is 5 nm - 5 micrometers

especially preferably 1 nm - 10 micrometers preferably. As a method of forming a luminous

layer, for example, \*\* can be formed by publicly known methods, such as the LB method, as

[ vacuum deposition, spin coat method, and cast method ], and it is preferred that it is especially
a molecule deposited film. A molecule deposited film means here the thin film which deposited
from the vapor phase state of the substance and was formed, and the film solidified from solution
states or a liquid phase state, and it can classify by the difference of condensation structure and
higher order structure, and the functional difference resulting from it with what is called a
molecule built up film formed by the LB method. This luminous layer can thin-film-ize the
liquid which melted binding agents, such as resin, in the solvent with the compound of the
above-mentioned formula (I) with a spin coat method etc., and can form it as indicated by

JP,S59-194393,A etc.

[0015] "the allylene group of the carbon numbers 6-20 which may be replaced" in Ar of formula (I) -- P-phenylene group, m-phenylene group, 4, and 4-biphenylene group, 1, 4-naphthylene group, and a terphenylene group -- or [0016]

[Chemical formula 6]

[0017] \*\*\*\*\*\*\*\*\*\*. With "the allylene group of the carbon numbers 6-20 which may be replaced" in Ar<sup>1</sup>. That by which low-grade alkyl groups, such as one or two methyl groups, and an ethyl group, are replaced is mentioned to P-phenylene group, m-phenylene group, a 4,4-biphenylene group, a 1,4-naphthylene group, terphenylene groups, or these bases. Especially, as for each of Ar and Ar<sup>1</sup>, from a viewpoint on acquisition of material, and composition, it is preferred that it is P-phenylene group.

[0018] As "a low-grade alkyl group which may be replaced" in R<sup>1</sup>, For example, the alkyl group of the straight chain which may be replaced with the halogen atom etc., or the carbon numbers 1-5 of branching is meant, for example, a methyl group, an ethyl group, n-propyl group, an isopropyl group, a butyl group, t-butyl group, a pentyl group, the Krol methyl group, the Krol ethyl group, etc. are mentioned. With "the heterocycle group of a five-membered ring or six

membered-rings", a furil group, a pyridyl group, an indolyl group, an indazolyl group, a PIRAZO lysyl-group, a pyrazinyl group, a quinolyl group, a pyrazolyl group, etc. are mentioned. With "the aralkyl group which may be replaced", benzyl, P-methylbenzyl group, P-Krol benzyl, a benzhydryl group, 2-naphthalene methylene group, 9-anthracene methylene group, etc. are mentioned. As "an aryl group of the carbon numbers 6-14 which may be replaced", A phenyl group, 4-biphenyl group, a naphthyl group, 2-anthryl group, P-methylene phenyl group, P-methoxypheny group, m-methylphenyl group, 1, 2-methylenedioxyphenyl group, 1-naphthyl group, 2-naphthyl group, etc. are mentioned. Especially, the phenyl group replaced by electron-donative [, such as a phenyl group or a methoxy group, a methyl group, or an ethyl group, ] is preferred.

[0019] As a "low-grade alkyl group" in  $R^2$  and  $R^3$ , the straight chain of the carbon numbers 1-5 or the alkyl group of branching is meant, for example, a methyl group, an ethyl group, n-propyl group, an iso-propyl group, a butyl group, t-butyl group, a pentyl group, etc. are mentioned. As "an aryl group of the carbon numbers 6-14 which may be replaced", the thing in  $R^1$  and the same thing can be used. both [ especially ]  $R^2$  and  $R^3$  -- although -- it is preferred that P-methoxypheny group or  $R^2$  is [ a methyl group and  $R^3$  ] a phenyl group.

[0020] The compound of following formula (II) is preferably mentioned among the compounds of formula (I) in this invention.

[0021]

#### [Chemical formula 7]

$$\begin{array}{c|c} & & & \\ \hline & &$$

[0022] (A hydrogen atom, a halogen atom, a low-grade alkyl group or a lower alkoxy group, and n of  $R^4$  are the integers of 1 or 2 among a formula.)

In the above-mentioned formula (II), as a "low-grade alkyl group" of R<sup>4</sup>, As the same low-grade alkyl group as the above-mentioned R<sup>2</sup>, and a "lower alkoxy group", the alkoxy group of the carbon numbers 1-5 is meant, a methoxy group, an ethoxy basis, a propoxy group, a butoxy group, and a pentoxy group are mentioned, and a methoxy group and an ethoxy basis are preferred especially.

[0023] The distilbene compound shown below is specifically preferred, and the illustration compounds 1, 6, 9, 12, 17, 18, and 32 which are compounds of formula (II) are more preferred especially.

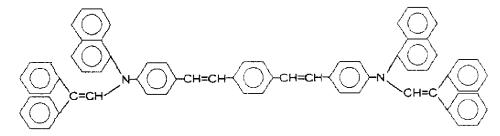
[0024]

[Chemical formula 8]

[0025]

[Chemical formula 9]

# 例示化合物7



#### 例示化合物8

# 例示化合物9

# 例示化合物10

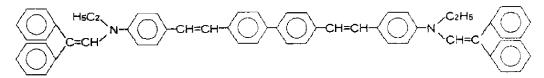
# [0026]

# [Chemical formula 10]

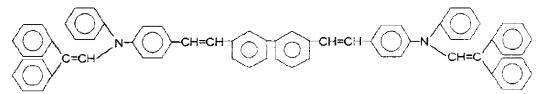
[0027]

# [Chemical formula 11]

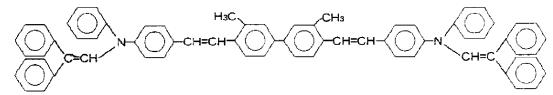
# 例示化合物17



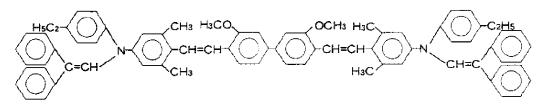
#### 例示化合物18



#### 例示化合物19



#### 例示化合物20



[0028]

# [Chemical formula 12]

# 例示化合物21 例示化合物22 例示化合物23 例示化合物24 例示化合物25

[0029]

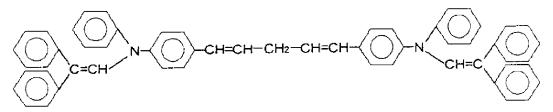
[Chemical formula 13]

# 例示化合物27

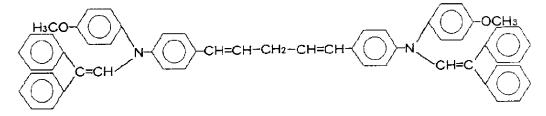
#### 例示化合物28

[0030]

[Chemical formula 14]



# 例示化合物30



# 例示化合物31

[0031]

[Chemical formula 15]

#### 例示化合物33

#### 例示化合物34

[0032] As for the glue line of the organic EL device in this invention, it is preferred to form using an adhesive high material to a luminous layer and an anode. As a material of a glue line, the metal complex of 8-hydroxyquinoline or its derivative can be mentioned. Specifically, it is a metal chelate oxy NOIDO compound containing the chelate of oxines, such as an eight quinolinol or 8-hydroxyquinoline. The thickness of the glue line in this case needs to be thinner than the thickness of the above-mentioned luminous layer, in order to maintain the luminescent color for a blue system, and it is 5-30 nm especially preferably 1-50 nm preferably. Although this glue line can be formed by publicly known methods, such as a spin coat method, the cast method, and vacuum deposition, for example, its vacuum deposition is preferred in as well as [ a

luminous layer and a hole injection layer ].

[0033] It is preferred to form as an anode of the organic EL device in this invention using electrode materials, such as small (4 eV or less) metal of a work function, an alloy, electrical conductivity compounds, and these mixtures. As an example of such an electrode material, sodium, a sodium potassium alloy, magnesium, lithium, magnesium / copper mixture, aluminum/(aluminum<sub>2</sub>O<sub>3</sub>), indium, etc. are mentioned. Although the thickness in particular of these electrode materials is not limited, it is preferred to choose suitably in 50-200 nm preferably 10 nm - 1 micrometer. When forming an anode, the above-mentioned electrode material can be produced by publicly known methods, such as vacuum evaporation and sputtering. As for the sheet resistance as an anode, it is preferred to set up below hundreds of ohms / \*\* become. In this EL element, it is preferred that either one of a cathode or an anode is transparent or translucent so that luminescence can be made to penetrate.

[0034] In the organic EL device of this invention, although it is not necessarily required, since it is improvement in luminescent ability, it is preferred to form a hole injection layer between a cathode and a luminous layer. The material which conveys an electron hole to a luminous layer by lower electric field as a material which forms this hole injection layer is preferred, and it is more preferred that the mobility of an electron hole is a  $10^{-6}$ cm<sup>2</sup> bolt and a second at least in the electric field of a  $10^{1}$  -  $10^{6}$  bolt / cm.

[0035] As a material which forms a hole injection layer, arbitrary things can be chosen and used

from the former out of what is commonly used as an electric charge pouring transported material of an electron hole in photoconductive material, or the publicly known thing used for the hole injection layer of an EL element, for example. As a former example, specifically A silazane derivative (US,4950950,B Description), A polysilane system (JP,H2-204996,A), an aniline system copolymer (JP,H2-282263,A), They are mentioned by conductive polymer oligomer, thiophene oligomer, etc. which were shown on the Tokuganheil-211399 Descriptions, and as a latter example, A triazole derivative (references, such as the US,3,112,197,B Description), An oxadiazole derivative (references, such as the US,3,189,447,B Description), an imidazole derivative (references, such as JP,S37-16096,B) and a poly aryl alkane derivative (the US,3,615,402,B Description.) The No. 3,820,989 Description, the No. 3,542,544 Description, JP,S45-555,B, A 51-10983 gazette, JP,S51-93224,A, a 55-17105 gazette, A 56-4148 gazette, a 55-108667 gazette, a 55-156953 gazette, Reference and pyrazoline derivatives, such as a 56-36656 gazette, and a pyrazolone derivative (the US,3,180,729,B Description, the No. 4,278,746 Description, JP,S55-88064,A) A 55-88065 gazette, a 49-105537 gazette, a 55-51086 gazette, A 56-80051 gazette, a 56-88141 gazette, a 57-45545 gazette, References, such as a 54-112637 gazette and a 55-74546 gazette, a phenylenediamine derivative (the US, 3,615,404,B Description.) JP,S51-10105,B, a 46-3712 gazette, a 47-25336 gazette, References, such as JP,S54-53435,A, a 54-110536 gazette, and a 54-119925 gazette, an arylamine derivative (the US,3,567,450,B Description and the No. 3,180,703 Description.) The No. 3,240,597 Description, the No. 3,658,520 Description, the No. 4,232,103 Description, The No. 4,175,961 Description, the No. 4,012,376 Description, JP,S49-35702,B, Reference, amino substitution chalcone derivatives (references, such as the 3,526,501st Description of an United States patent), such as a 39-27577 gazette, JP,S55-144250,A, a 56-119132 gazette, a 56-22437 gazette, and the West German patent No. 1,110,518 Description, An oxazole derivative (thing given in the US,3,257,203,B Description etc.), A styryl anthracene derivative (references, such as JP,S56-46234,A), a fluorenone derivative (refer to JP,S54-110837,A) and a hydrazone derivative (the US,3,717,462,B Description.) JP,S54-59143,A, a 55-52063 gazette, a 55-52064 gazette, A 55-46760 gazette, a 55-85495 gazette, a 57-11350 gazette, References, such as a 57-148749 gazette and JP,H2-311591,A, a stilbene derivative (JP,S61-210363,A and a 61-228451 gazette.) A 61-14642 gazette, a 61-72255 gazette, a 62-47646 gazette, References, such as a 62-36674 gazette, a 62-10652 gazette, a 62-30255 gazette, a 60-93445 gazette, a 60-94462 gazette, a 60-174749 gazette, and a 60-175052 gazette, etc. can be mentioned.

[0036] A hole injection layer can be formed by, for example, forming membranes by publicly known methods, such as a vacuum deposition method, a spin coat method, the cast method, and the LB method, as two or more lamination layers depended on the monolayer by one sort of the above-mentioned compound, the monolayer by two or more sorts of combination, or two sorts or more. Although the thickness in particular as a hole injection layer in this case is not limited, 5 nm - 5 micrometers are preferred.

[0037] Next, the electrophotographic photoreceptor in this invention is explained. As a conductive substrate in the electrophotographic photoreceptor of this invention, The photosensitive layer which besides consists of a charge generating layer and a charge transport layer is formed, an electric field is applied to a photosensitive layer, it is for moving the carrier (an electron or an electron hole) refined by the optical absorption of the photosensitive layer, and the conductor of the form of a drum, a plate, a sheet, etc. can be used. As for the material used for a conductor, paper or a plastic etc. which performed metal, such as copper and aluminum, carbon, and conductive processing, for example is mentioned.

[0038] On the conductive substrate in the electrophotographic photoreceptor of this invention, the photosensitive layer which consists of a charge generating layer and a charge transport layer is formed. As a charge generating layer, screw azo compounds, such as clo RODAI Ambur, a jib -- materials, such as polycyclic quinone system compounds, such as ROM anthanthrone, a perylene system compound, a quinacridone series compound, a phthalocyanine system compound, and an AZURENIUMU salt system compound, -- one sort -- or two or more sorts can be used together and it can form. It is vacuum deposition, and the above-mentioned charge generating layer material is distributed using the method of forming a film with a direct compound, and a thermoplastic high polymer binder, and methods of forming a charge generating layer using such materials include the method of applying and forming membranes, etc. As for the thickness of the charge generating layer in this case, about 0.1-1.0 micrometer is

preferred.

[0039] As a charge transport layer in the electrophotographic photoreceptor of this invention, it is preferred to use the distilbene compound of the above-mentioned formula (I). That is, the substance generally used for a charge transport layer, Especially a molecular weight has a comparatively large molecular weight among about 200 to 800 low molecular weight compounds, And it is because excelling in respect of performances, such as the aromatic amine compound which has a share double bond being unable to oxidize easily with oxidizing gases, such as stability, sensitivity of a photo conductor, and ozone, and having a fixed mechanical strength, is known. If the distilbene compound of the above-mentioned formula (I) is independent, since it is deficient in film forming ability, it is preferred to form tales doses, in addition a charge transport layer for a thermoplastic high polymer binder mostly. As a thermoplastic high polymer binder in this case, ester system resin, such as polycarbonate resin, polyarylate resin, and polyester resin, can be used. Although the thickness in particular of a charge transport layer is not limited and can be suitably chosen according to a situation, it is 5 micrometers - 30 micrometers preferably. Formula (III) among the distilbene compounds of formula (I) used for this charge transport layer [0040]

[Chemical formula 16]

[0041] (Ar<sup>2</sup> expresses among a formula a methylene group or a phenylene group, the low-grade alkyl group by which R<sup>5</sup> may be replaced, the aralkyl group which may be replaced, or the aryl group of the carbon numbers 6-14 which may be replaced.) -- the compound shown is preferred. What has "the low-grade alkyl group which may be replaced, the aralkyl group which may be replaced, or the aryl group of the carbon numbers 6-14 which may be replaced" be [ the same as that of what is defined by R<sup>1</sup> of the above-mentioned formula (I) ] it in R<sup>5</sup> of formula (III) is mentioned.

[0042] when using the distilbene compound of formula (I) for an electrophotographic photoreceptor element, in order to dissolve in an organic solvent and to use these distilbene compound in many cases, solubility is usually bad — the distilbene compound of a high-melting point is not comparatively preferred. As an organic solvent in this case, ketone, such as acetone and methyl ethyl ketone. Aprotic polar solvents, such as aromatic hydrocarbon, such as ether, such as ester species, such as ethyl acetate and butyl acetate, a tetrahydrofuran, and dioxane, benzene, and toluene, N.N-dimethylformamide, and dimethyl sulfoxide, etc. are mentioned. As a soluble good compound, Ar in the compound of formula (I) A methylene group, They are m-phenylene group, a 3,5-dimethyl- P-phenylene group, etc., As for each of phenyl groups by which R¹ was replaced by the low-grade alkyl group and the electron donative group, R², and R³, the compound whose methyl group and another side any one of a phenyl group or R², and the R³

is phenyl groups is mentioned.

[0043] Specifically, the illustration compounds 1, 7, 9, 12, 29, and 30 which are compounds of formula (III) are more preferred especially among the following compounds shown as a distilbene compound of formula (I). The new distilbene compound used for this invention is easily compoundable according to the following synthetic pathway.

[0044] That is, formula (IV)

[0045]

[Chemical formula 17]

$$R^1-N-Ar^1-CH=CH-Ar-CH=CH-Ar^1-N-R^1$$
 (IV)

[0046] (the sign is synonymous with formula (I) among a formula.) -- the compound shown -- formula (V)

[0047]

[Chemical formula 18]

$$H-C-CH$$
 $R^2$  $(v)$ 

[0048] (the sign is synonymous with formula (I) among a formula.) -- the compound shown and the mole ratio of 1:2 to 1:2.5 -- benzene. It is easily compoundable by making it react into solvents, such as mono- chlorobenzene, ethyl acetate, toluene, or ethanol, adding a small amount

of P-toluenesulfonic acid, naphthalene sulfonic acid, chlorides, or acetic acid further, and removing the water heated and generated out of a system.

[0049] A new distilbene compound is compoundable also according to another synthetic pathway. Namely, formula (VI)

[0050]

[Chemical formula 19]

$$\begin{array}{ccc}
R^{2} & R^{1} \\
R^{3} & C = CH & N - Ar^{1} - CHO
\end{array}$$
(VI)

[0051] (the sign is synonymous with formula (I) among a formula.) -- the compound shown -- formula (VII) [0052]

[Chemical formula 20]

[0053] (R of the above, homonymy, and other signs is synonymous with formula (I) among a formula.) -- it being the compound and the mole ratio of 2:1 to 2.5:1 which are shown, and, Dimethylformamide, dimethylsulfo oxide, dimethylacetamide, making it react in solvents, such as ethanol, -- further -- sodium ethoxide, potassium tert-butoxide, etc. -- the compound of formula (VII) -- almost -- a \*\*mol -- in addition, it is easily compoundable by making it react under a room temperature or 70-100 \*\* temperature up.

145.5 \*\*.

[Working example] Below, the embodiment of this invention is described.

The synthetic example 1 (composition of the illustration compound 1)

1-a) Dissolve 32g of synthetic diphenylamine and 36.0 g of diphenylacetaldehyde of 1,1diphenyl-2-(diphenylamino) ethylene in about 400 ml of benzene. 20 mg of p-toluenesulfonic acid is added to this, it heats, and the water produced by a reaction is excluded out of a system. After removing benzene etc., the white powder which \*\*\*\*(ed) is recrystallized from ethyl acetate, and the white crystals 56g are obtained. The melting point was 149.0-150.0 \*\*. 1-b) The enamine compounds 10g compounded by synthetic 1-a of the monoaldehyde object of 1,1-diphenyl-2-(diphenylamino) ethylene are dissolved in 20 ml of dichloroethanes, After adding this under ice-cooling gradually into 8.9 g (5.4 ml) of phosphorus oxychloride, and the BIRUSU Meyer's reagent prepared by 5.6 g of dimethylformamide, it reacts by raising reaction temperature gradually the bottom of ice-cooling, a room temperature, and heating (internal temperature of 80-85 \*\*). After adding this reaction mixture to 5 N-NaOH underwater and making reaction mixture into alkalescence, the object dissolved into the dichloroethane is taken out and the powdered crystal 9.1g of yellowish white is obtained. The melting point was 145.0-

[0055] As a result of measuring IR of this compound (KBr tablet method), absorption of the aldehyde group was accepted near 1730 cm<sup>-1</sup>.

xylene, and 11 g of phosphorous acid triethyl of p-phenylene group content phosphorus compounds under a nitrogen air current and in the oil bath (temperature of 130 \*\*) for 6 hours. Next, decompression distilling off of superfluous phosphorous acid Tori Eril and the subgenerated ethyl chloride was carried out. If this is neglected in ether, the white powder 7.2g will be obtained. The melting points were 68.5-70.0g.

1-c) Heating and stirring was performed for the composition alpha, 4.5 g of alpha'-dichloro p-

1-d) The Wittig compound 1.3g obtained by synthetic 1-c of the illustration compound 1 and the aldehyde compound 5.0g obtained by 1-b were dissolved in 50 ml of dimethylformamide, 1.5 g of tert-butoxy potassium was gradually added to this from Spa Zillah under the room temperature, and churning was performed for 2 hours. The yellow powder which has the strong fluorescence which deposited is taken out, subsequently heat acetone washes enough, and a 2.3-g N and N'-JIENAMIN group content distilbene compound is obtained. The melting point was 248.5-250.5 \*\*.

[0056] As a result of measuring IR of this compound (KBr tablet method), the absorption based on methylidyne was observed in 1520 cm<sup>-1</sup> and 1620 cm<sup>-1</sup>.

The synthetic example 2 (composition of the illustration compound 6)

2-a) Dissolve 20g of synthetic phenyl-(4-methoxypheny) amine and 21 g of diphenylacetaldehyde of 1-phenyl-1-(4'-methoxypheny)-2-(diphenylamino) ethylene in 300 ml of benzene. 15 mg of p-toluenesulfonic acid is added to this, it heats, and the water produced by

a reaction is excluded out of a system. If heating flowing back is performed in 100 ml of after-removal methanol, white powder deposits benzene etc. this powder -- ethyl acetate: -- from methanol, recrystallization is performed and the white crystal 23.4g is obtained. The melting point was 118-120 \*\*.

2-b) The enamine compounds 12g compounded by synthetic 2-a of the monoaldehyde object of 1-phenyl (4-methoxypheny)-2-(diphenylamino) ethylene are dissolved in 20 ml of dichloroethane, After adding this under ice-cooling gradually into 8.9 g (5.4 ml) of phosphorus oxychloride, and the BIRUSU Meyer's reagent prepared by 6.5 g of dimethylformamide, it reacted under ice-cooling by raising reaction temperature gradually subsequently the bottom of a room temperature and heating (internal temperature of 80-85 \*\*), and going. After adding reaction mixture into a 5N-NaOH aqueous solution finally and making a water layer into alkalescence, the object dissolved into the dichloroethane is taken out, from ethyl acetate, recrystallization is performed and the white powdered crystal 9.4g is obtained. The melting points were 112.5 \*\* - 114.0 \*\*.

2-c) p-phenylene group content phosphorus compounds 2.1g compounded by synthetic 1-c of the illustration compound 6 and the monoaldehyde object 9.0g compounded by 2-b are dissolved in 100 ml of dimethylformamide, 2.5 g of tert-butoxy potassium was gradually added to this from Spa Zillah under the room temperature, and churning was performed for 2 hours. The yellow powder which has the strong fluorescence which deposited is taken out, subsequently acetone

washes enough, and a 4.1-g N and N'-JIENAMIN group content JISUTERUBEN compound is obtained. The melting point was 234.5-236.5 \*\*.

[0057] The absorption based on a methylidyne group was accepted near 1520 cm<sup>-1</sup> and 1620 cm<sup>-1</sup> like [ IR measurement of this compound ] the illustration compound 1.

The synthetic example 3 (composition of the illustration compound 3)

3-a) Dissolve 32g of synthetic diphenylamine and 42 g of JI (p-methoxypheny) acetaldehyde of 1,1-diphenyl-2-JI (p-methoxypheny) aminoethylene in about 500 ml of benzene. 20 mg of p-toluenesulfonic acid [ 20 mg of ] is added to this, and the water produced with the Dean Stark pipe as a result of a reaction is excluded out of a system. After removing benzene etc., methanol washes the white powder which \*\*\*\*(ed), it recrystallizes from ethyl acetate, and the white crystals 389g are obtained.

3-b) Obtain the crystal of the yellowish white shown in a following formula with the same synthesizing method as synthetic 1-b of the monoaldehyde object of 1,1-diphenyl-2-JI (p-methoxypheny) aminoethylene. The melting point was 125.5-127.0 \*\*.

[0058]

[Chemical formula 21]

[0059] 3-c) Obtain the yellow-orange color crystal which has strong fluorescence with the same synthesizing method as synthetic 1-d of the illustration compound 3. Heat washing by dimethylformamide refined the object. The melting point was 254.5-257.0 \*\*. The absorption based on a methylidyne group was accepted near 1525 cm<sup>-1</sup> and 1630 cm<sup>-1</sup> like [ IR measurement of this compound ] the illustration compound 1.

The synthetic example 4 (composition of the illustration compound 12)

4-a) Heating flowing back was performed for the synthetic 4,4'-bis(KURORU methyl)biphenyl 7.0g and 1 g of phosphorous acid triethyl of biphenylene group content phosphorus compounds the bottom of an N<sub>2</sub> air current, and among the oil bath for 5 hours.

[0060] After carrying out decompression distilling off of superfluous phosphorous acid triethyl, the ethyl chloride of a by-product, etc., the depositing white crystals 8.4g of the following formula were obtained. The melting point was 98-100.5 \*\*.

[0061]

[Chemical formula 22]

[0062] This compound showed negative in the by RUSHUTA yne test, and it turned out that halogen is not contained.

4-b) The Wittig compound 4.2g obtained by synthetic 4-a of the illustration compound 12 and

the monoaldehyde object 7.5g acquired by 1-b were dissolved in 50 ml of dimethylformamide, 2.4 g of potassium tert-butoxide was gradually added to this, and it agitated under the room temperature for 4 hours. The orange powder which has the strong fluorescence which deposited was taken out, enough, after rinsing, heat acetone washed enough and the orange powder 6.5g was obtained. The melting point was 287.5-289.5 \*\*.

The synthetic example 5 (composition of the illustration compound 14)

The Wittig compound 4.2g obtained by 4-a and the monoaldehyde object 8.3g acquired by 3-b were dissolved in 50 ml of dimethylformamide, 2.4 g of potassium tert-butoxide was gradually added to this, and it agitated under the room temperature for 4 hours. The orange powder which has the strong fluorescence which deposited was taken out, enough, after rinsing, heat acetone washed and the strong orange fluorescent substance 6.7g was obtained. The melting point was 293.5-295.5 \*\*.

The synthetic example 6 (composition of the illustration compound 29)

6-a) Heating flowing back of 10 g of synthetic 1,3-dibromopropane and 20 g of phosphorous acid triethyl of methylene group content phosphorus compounds was carried out under an  $N_2$  air current and in the oil bath for 5 hours. The oily water-white substance 7g of the following formula was obtained by carrying out decompression distilling off of the superfluous phosphorous acid triethyl etc., and also carrying out decompression refining.

[Chemical formula 23]

[0064] 6-b) The Wittig compound 2.8g obtained by synthetic 6-a of the illustration compound 29 and the monoaldehyde object 8.0g acquired by 1-b were dissolved in 20 ml of dimethylformamide, the alcohol solution of sodium ethylate was gradually added to this, and churning was performed for bottom 4 hours of a room temperature. The candy-like object thing which added 100 ml of water after neglect and into reaction mixture overnight [ back ], and deposited was extracted from 50 ml of ethyl acetate, and it dried enough. Then, if ethyl acetate is distilled off and heating flowing back of the residue is carried out in 50 ml of methanol, yellowish white powder will generate gradually. The generated powder is taken out and it separates from an unreacted monoaldehyde object by carrying out column separation by benzene:n-hexane.

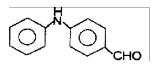
[0065] The strong powder of the yellow taste which has 3.6 g of fluorescence was obtained after \*\*\*\* with methanol. The melting point was 145.5-147.0 \*\*.

The synthetic example 7 (composition by the exception method of the illustration compound 1)
7-a) Dissolve 40 g of aldehyde of synthetic N-allyldiphenylamine of the monoaldehyde object of diphenylamine in 400 ml of ethyl acetate, add 100 ml of 1N-hydrochloric acid aqueous solutions to this, and agitate violently under a room temperature for 2 hours. An ethyl acetate layer

changes from a yellow solution to a green solution with hydrolysis. Subsequently, after drying an ethyl acetate layer from sulfuric anhydride soda, ethyl acetate is distilled off, a residue is recrystallized from ether, and the green and white powder 26g of a following formula is obtained. The melting point was 52.0-54.5 \*\*.

[0066]

#### [Chemical formula 24]



[0067] 7-b) Obtain the distilbene compound 4.2g for the monoaldehyde object 4.0g of synthetic 7-a of the illustration compound 1, and the content phosphorus compounds 2.1g compounded by 1-c like 2-c. The diphenylacetaldehyde of a mol is added to this distilbene compound 2.2 times, and the illustration compound 1 is obtained by adding and forming the acetic anhydride of a small quantity in dichlorobenzene into heating drying.

[0068] Although a little compounds of the synthetic example 1 and compounds of the synthetic example 7 were mixed every, the capillary tube was stuffed and the fall phenomenon of the melting point was observed, the fall phenomenon was not accepted at all. Although IR measurement was performed, the spectrum was completely the same.

As shown in Embodiment 1 - 5 organic-electroluminescence-element <u>drawing 1</u>, what vapordeposited ITO12 by a thickness of 0.1 micro was used as a transparent supporting board on the 1.1-mm-thick glass substrate 11. Subsequently this transparent supporting board was cleaned ultrasonically in pure water among isopropyl alcohol, and the last UV ozone wash was performed.

[0069] This glass substrate 11 is fixed to the electrode holder of an evaporation apparatus (product made from Japanese Vacuum technology), and it is N,N'-bis(3-methylphenyl)-N,N'diphenyl to the resistance heating boat made from molybdenum. [1,1'-biphenyl] (TPD) 200 mg was put in, 200 mg of illustration compounds 1, 3, 6, 12, and 22 shown in Table 1 were put into another board made from molybdenum, it ranked second and the vacuum chamber was decompressed to 1x10<sup>-4</sup>Pa (ionization energy: 5.85 eV). Then, said board containing TPD was heated to 215-220 \*\*, TPD was vapor-deposited on ITO12 with the evaporation rate of 1-0.3nm/sec, and the hole injection layer 13 of 0.06 micro of thickness was formed. [0070] Then, the luminous layer 14 which has 0.04 micrometer of thickness was formed by lamination vacuum evaporation from the boat by which the illustration compounds 1, 3, 6, 12, and 22 shown in Table 1 entered on the hole injection layer 13, without taking out the obtained glass substrate 11 from a vacuum chamber. As for vacuum evaporation conditions, the boat temperature of 0.1-0.3nm/[ sec and ] and substrate temperature of 330 \*\* and an evaporation rate was a room temperature.

[0071] Subsequently, the obtained glass substrate 11 was taken out from the vacuum chamber, the mask made from a stainless steel was installed on the luminous layer 14, and it fixed to the

substrate holder again. Next, 200 mg of tris(eight quinolinol) aluminum (Alq<sub>3</sub>) was put into the board made from molybdenum, and it laid in the vacuum chamber. The magnesium ribbon 1g was put into another boat made from molybdenum, and 500 mg of silver wires were put into another basket made from tungsten. Then, after decompressing a vacuum chamber to 1x10<sup>-4</sup>Pa, while heating the boat containing Alq<sub>3</sub> to 230 \*\* and vapor-depositing 0.02micro Alq<sub>3</sub> on the luminous layer 14 with the evaporation rate of 0.01 - 0.03 nm/s, Simultaneously, silver was vapor-deposited with the evaporation rate of 0.1 nm/s, and magnesium was vapor-deposited with the evaporation rate of 1.4 nm/s from another boat made from molybdenum with the resistance heating method. 1.5-micrometer lamination vacuum evaporation of the mixed metal electrode of magnesium and silver was carried out on the luminous layer on the above-mentioned conditions, and the anode 15 was formed.

[0072] Thus, the half line of the maximum light emitting luminance when the voltage of 15V is impressed between the cathode 12 of an organic electroluminescence element and the anode 15 which were produced, and the maximum light emitting luminance was measured. The result was shown in Table 1. Data are placed into a vacuum and the data of Table 1 measures them.

[0073]

[Table 1]

実施例	発色体材料	4	最大発光輝度(cd/m²)	半減時間(分)	
1	例示化合物	1	4400	120	
2	"	3	6000	90	
3	"	6	5600	280	
4	"	12	7800	110	
5	"	22	9600	160	

[0074] Embodiment 6-10 The aluminum film 22 of 10 micrometers of thickness was vapordeposited on the 75-micrometer polyester film 21 of thickness as the electrophotographic photoreceptor element conductive substrate 23. 5 g of oxy titanium phthalocyanine is added to 100 g of dimethoxyethane, In addition to the solution which dissolved 3 g of polyvinyl butyral resin (the DENKI KAGAKU KOGYO [ K.K. ] make, a trade name: electrification butyral \*\*6000C) in 50 g of dimethoxyethane, dispersion liquid were obtained after carrying out SANDOGU lined processing. On the above-mentioned conductive substrate 23, this liquid was applied so that the thickness after desiccation might be set to 0.4 micrometer, and the charge generating layer 24 was formed.

[0075] The illustration compounds 1, 2, 9, 24, and 29 of Table 2 as a charge transport nature substance 10 g each, The solution which dissolved 10g of poly car PONETO resin (Mitsubishi Gas Chemical, Z-400) and 0.2 g of vitamin E in 100 ml of dioxane was prepared, on the charge generating layer 24, this solution was applied so that the thickness after desiccation might be set to 23 micrometers, the charge transport layer 25 was formed, and the sample was created.

[0076] Thus, the electrophotographic properties of the created sample were evaluated using electrostatography paper test equipment (the product made from the Kawaguchi electrical and electric equipment, SP-428). The result is shown in Table 2. The electrification pressure of Table 2 is the potential at the time of carrying out negative electrification by impressing voltage so that corona current may be set to 22microA.

Sensitivity is the electric discharge light exposure which the surface voltage at the time of irradiating with 1.0 lux white light required for halving from -500V to -250V, and rest potential is the potential 10 seconds after exposure.

The electrophotographic properties of 10,000 times after are the electrification pressure and rest potential of a \*\*\*\*\*\*\* case about 10,000 empty aging examinations using the equipment (light exposure: 20 lux x 1 second).

[0077]

[Table 2]

実施例	電荷輸送性 化合物			初期特性	1万回後の特性		
天心时			帯電圧 (V)	感度 E1/2(luxsec)	残留電位 (V)	<b>帯電圧</b> (V)	残留電位 (V)
6	例示化合物	1	-710	0.12	0	-685	-3
7	"	2	-705	0.11	0	-685	-5
8	"	3	-680	0. 15	0	-640	-3
9	"	4	-715	0.10	0	-695	-14
10	"	5	-640	0.10	0	-625	-5

[0078]

[Effect of the Invention] The outstanding performance is shown when the distilbene compound which has a specific new structure is used for the luminous layer of the organic electroluminescence element of this invention, and the charge transport layer of an electrophotographic photoreceptor element. That is, when it uses as an organic electroluminescence element, the long lasting photogen by highly efficient nature, thermal stability, and high amorphism nature, such as high-intensity and a well head, can be provided.

[0079] When it uses as an electrophotographic photoreceptor element, it has the outstanding hole transport efficiency (- 10<sup>-4</sup>cm<sup>2</sup>/V), and high sensitivity with little photodegradation, thermal degradation, and mechanical deterioration and the photo conductor for \*\*-proof [ high ] can be provided.

[Brief Description of the Drawings]

[Drawing 1] It is an outline sectional view showing the embodiment of the organic electroluminescence element of this invention.

[Drawing 2] It is an outline sectional view showing the embodiment of the electrophotographic photoreceptor of this invention.

[Explanations of letters or numerals]

11 Glass substrate

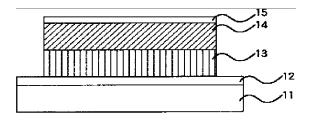
12 ITO (cathode)

13 Hole injection layer

- 14 Luminous layer
- 15 Metal electrode layer
- 21 Polyester film
- 22 Aluminum film
- 23 Charge generating layer
- 24 Charge transport layer

# [DRAWINGS]

# [Drawing 1]



# [Drawing 2]

